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10-12-2019

Flexnode Plus Project:
Results of testing a hydrogen-based reversible fuel cell system

1. Abstract

Within the Flexnode Plus project the long-term degradation characteristics of a proton exchange membrane (PEM) electrolyzer (5.5 kW, AC, 1 Nm^3/h H_2) and fuel cell (1.0 kW, DC, 0.9 Nm^3/h) was experimentally tested. The electrolyzer unit was operated at various loads and pressures for approximately 750 hours in total, while the fuel cell was operated at a constant load of 1 Ω resistance for approximately 1120 hours in total. The efficiency of the hydrogen production in the electrolyzer and the electricity production in the fuel cell was expressed using the hourly average system efficiency and average cell efficiency. In order to predict the state of health and remaining lifetime of the electrolyzer cell and fuel cell, the decay of the cell voltage over time was monitored and the direct mapping from aging data method was used.
The electrolyzer cell showed a stable cell voltage and cell efficiency in the studied time period, with an average cell voltage decay rate of 0.5 µV/h. The average cell voltage of the fuel cell dropped with a rate of 2 µV/h during the studied time period.

2. Project background

Flexnode Plus is a project initiated by GasTerra, GasUnie and Hanze University of Applied Sciences, Centre of Expertise Energy (Hanze). The project studies the main physical and the long-term degradation characteristics of a PEM electrolyzer and fuel cell, that together form a reversible hydrogen-based fuel cell system. The experimental setup has been acquired and built during the previously completed RVO project: Flexnode (TESI115003)[1]. As the experimental setup was already installed and operational, further measurements could be carried out at a relatively low cost. This project aims to use the already existing experimental setup to further test the potential of hydrogen as a green energy carrier within the water electrolysis-to-hydrogen-to-electricity cycle.

This report presents the main findings and results of the experiments carried out at the testing site facility of Hanze (EnTranCe), in the period of January - December 2019.

3. A brief theory of PEM electrolyzers and fuels cells

A proton exchange membrane (PEM) water electrolyzer (and fuel cell) is an alternative to the alkaline electrolyzer that uses extra pure water and has a solid polymer electrolyte between the anode and cathode. The polymer electrolyte membrane serves as a medium to transport the charge carrier between the anodic and cathodic compartments (see **Figure *1***). In the case of a PEM system, this charge carrier is a proton or, in other words, a positively charged hydrogen ion. Additionally, the membrane functions as a medium for the reduction of gas crossover, it keeps the system compact and supports the operation of the system at high pressure.[2]

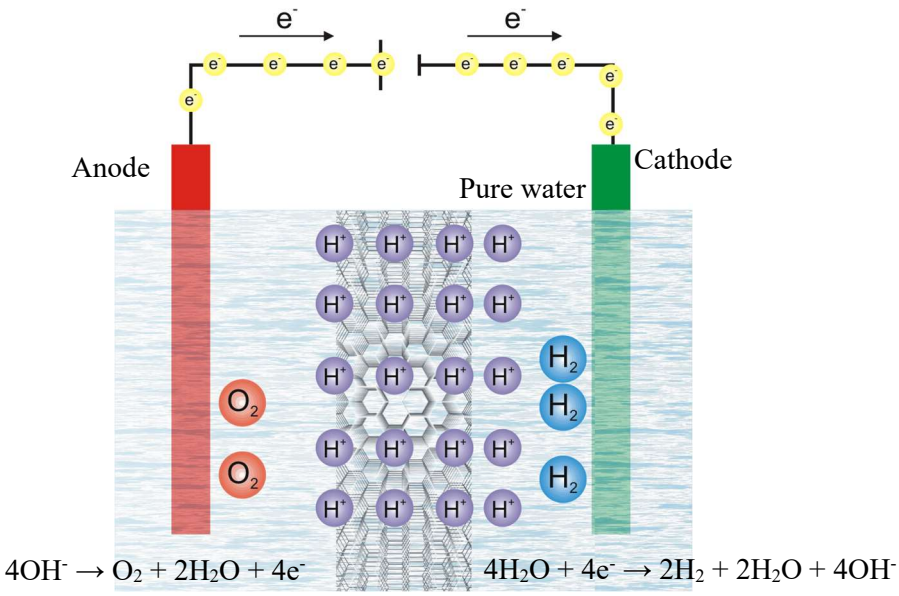

Figure 1. Conceptual representation of a PEM electrolyzer cell.

An electrochemical reaction is considered spontaneous if the sum of the reduction and oxidation standard potentials is positive. In the case of electrolysis this is clearly a negative number:

$$4H^+ + 4e^- \rightarrow + 2H_2 \qquad E^0_{Reduction} = 0V$$

$$\underline{2H_2O \rightarrow 4H^+ + 4e^- + O_2} \qquad \underline{E^0_{Oxidation} = -1.23V}$$

$$2H_2O \rightarrow O_2 + 2H_2 \qquad E^0_{Reaction} = -1.23V$$

By definition the reduction of the protons is set to be a standard potential of 0 V.

$$E^o_{reaction} = \frac{\Delta G_{dissociation}}{n \cdot F} = -1.23\ V$$

The minimum voltage difference between the anode and cathode at which a water molecule breaks down is 1.23 V. However, for low temperature electrolyzers (below 100 °C), the heat required for the reaction is also generated by the electric current. Therefore, the electrolyzer must be supplied with a voltage higher than the minimum value, due to the heat demand associated with the change in entropy and temperature.[3] This minimum voltage which also delivers the required minimum amount of heat is called thermoneutral voltage.

$$E^{o}_{Thermoneutral} = \frac{\Delta H_{dissociation}}{n \cdot F} = -1.48\ V$$

A low temperature electrolyzer that is able to generate hydrogen at the thermoneutral voltage is operating virtually at a 100% efficiency. The thermoneutral voltage is used in this report to calculate the cell efficiency of the tested electrolyzer.

In case of the PEM fuel cell, the same standard reaction voltages are applied, but with the opposite sign. The positive standard reaction voltage of 1.23 V and thermoneutral voltage of 1.48 V shows that the combination of hydrogen with oxygen is spontaneous. In this report, the thermoneutral voltage is also used for the cell efficiency calculations of the fuel cell.

4. The experimental setup

The experimental setup consists of the following main components:

1) ***Electrolyser (GreenHydrogen HyProvide P1)***:
- Nominal power: 5.5 kW, AC
- Max. hydrogen production rate: 1 Nm^3/h
- Max. H2 stack pressure: 50 bar
- 33 cells

2) ***Hydrogen storage tank (Mahytec, type IV tank)***:
- Storage volume: 850 Liters
- Max. storage pressure: 60 bar
- Polymer liner reinforced with composite material

3) ***Fuel cell (H2SYS AIRCELL® 1000 ACS)***:
- Nominal production power: 1.0 kW, DC
- Maximal power: 1.2 kW, DC (for max. 60 seconds)
- Max. Hydrogen consumption : 0.9 Nm^3/h
- 28 cells

The schematic of the complete reversible fuel cell system is shown in **Figure *2***.

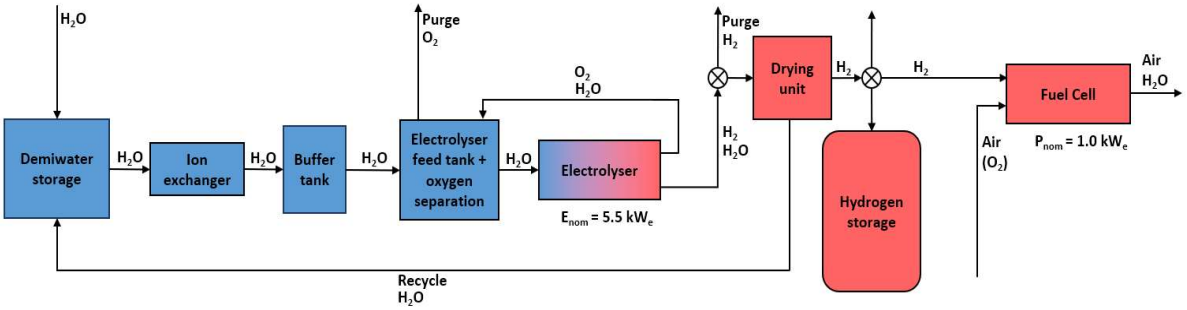

Figure 2. Schematic representation of the Flexnode experimental setup. (source: Marvin Bosker)

5. Methodology

Experimental procedure

The electrolyzer unit was operated at various loads and cell pressures for a total of about 750 hours to investigate long-term stability. 100 %, 75%, 50 % and 25 % of the nominal load were tested which corresponds to a current density of 1, 0.75, 0.5 and 0.25 A/cm^2, respectively. The operating temperature was set automatically by the electronics of the (commercially available) electrolyzer to 70 ºC and could not be changed. The produced hydrogen flow was measured with a Bronkhorst, type: F-111BI-20K mass flowmeter. The consumed electricity was monitored with an iEM3200 Schneider Electric energy meter.

The fuel cell was operated at a constant load of 1 Ω resistance for a total of about 1120 hours to investigate long-term stability. An automated load variation could not be achieved, therefore the load was varied manually only when the fuel cell was turned on or off. The constant 1 Ω electric load was provided by an AC/DC Chroma 63800 Series electronic load at DC, constant resistance mode. The 1 Ω resistance corresponds to 50 % of the nominal load, thus around 500 W. The consumed hydrogen flow was measured with a Bronkhorst, type: F-111BI-20K mass flowmeter. The cell voltage and current intensity was monitored by the built-in electronics of the fuel cell. The fuel cell was always operated at a constant temperature of around 40 ºC and at a constant hydrogen pressure of 1.5 bar.

All the measured data was collected and recorded using a homemade LabView program.

Calculations

The efficiency of the hydrogen production in the electrolyzer was expressed using system efficiency and average cell efficiency. These were calculated on an hourly basis. The cells are connected in series, therefore the average cell voltage was obtained by dividing the average stack voltage with the number of cells.

The system efficiency of the electrolyzer was calculated using the higher heating value energy-content of the produced hydrogen and the total electric energy consumption of the electrolyzer system:

$$\eta_{system\ ,electrolysis} = \frac{Energy\ out_{in\ form\ of\ H2,HHV}}{Energy\ in_{electricity}} \times 100$$

The average hourly hydrogen flow rate was used to calculate the amount of produced hydrogen.

The cell efficiency of the electrolyzer was expressed using the theoretical thermoneutral voltage and the measured average hourly cell voltage:

$$\eta_{cell} = \frac{Thermoneutral\ cell\ voltage_{theoretical}}{Cell\ voltage_{real}} \times 100$$

Similarly, the efficiency of the fuel cell was expressed using system efficiency and average cell efficiency. These were also calculated on an hourly basis.

The system efficiency of the fuel cell was expressed using the energy content (higher heating value) of the consumed hydrogen and the produced electric energy:

$$\eta_{system\ ,fuel\ cell} = \frac{Energy\ out_{electricity}}{Energy\ in_{in\ form\ of\ H2,HHV}} \times 100$$

The cell efficiency for the fuel cell was also calculated using the thermoneutral voltage:

$$\eta_{cell\ ,FC} = \frac{Cell\ voltage_{real}}{Thermoneutral\ cell\ voltage_{theoretical}} \times 100$$

Cell decay prognosis

In order to predict the state of health and remaining lifetime of an electrolyzer cell or fuel cell, the decay of the cell voltage over time is commonly used as a degradation characteristic.[4] Several prognostic approaches for the remaining lifetime are available using the cell voltage decay. Generally, these methods can be divided into two categories: (i) model based and (ii) data driven approaches:

(i) The model based approaches (physical model, system state observer-based method) uses complex electrochemical and physical equations to describe the actual

degradation process. It is out of the scope of this project to build a reliable physical and electrochemical model for lifetime prognosis.

(ii) Data-driven approaches (direct mapping from aging data, machine learning approaches, signal processing based method, statistical and probabilistic analysis) use measurement data and offer a higher flexibility.[4]

Machine learning approaches and statistical analysis need larger amounts of data than those available in this project. The direct mapping from aging data is a simple and straightforward linear fitting method. Although, it is not suitable to describe nonlinear behavior, taking in account the somewhat limited amount of data that could be gather during this project, the direct mapping with linear fitting method is used in this work to prognose cell decay.

6. Results and discussion

PEM electrolyzer

The average cell efficiency of the electrolyzer did not change significantly in the studied time period. **Figure *3*** shows the average hourly cell efficiency as a function of the operating time. It can clearly be seen, that at lower loads and current densities the stack is more efficient. At a 25 % load the cell efficiency is reaching almost 90 %, while at 100 % load the cell efficiency is staying steadily at around 80 %.

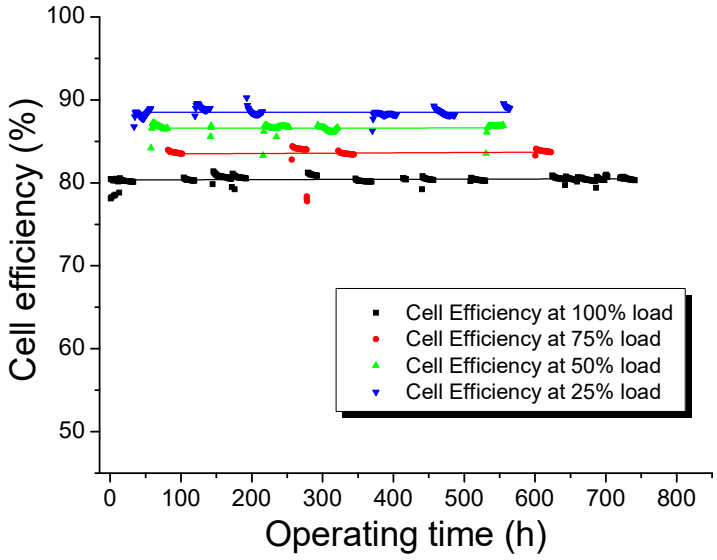

Figure 3. The calculated average hourly electrolyzer cell efficiencies shown as a function of the operating time.

The average cell voltage at a 100 % load was used to prognose the degradation rate and estimate the expected cell lifetime. It can be noticed on ***Figure 4*** that during the several hours long measurements, the cell voltage increases gradually and then asymptotically reaches a plateau. An increased cell voltage means a lower efficiency. During the non-

operating time between the measurements however, the cell voltage is regenerated. This behaviour is probably caused by the proton depletion of the PEM membrane and the limited diffusion rate of the newly forming hydrogen ions. The resistance of the membrane is controlled by the proton flow-rate, and therefore this also influences the cell voltage.
There are also some differences between the stack pressures from measurement to measurement, but the effect of the pressure was found to be minimal, and it is ignored. In a real life scenario the operating pressure of the electrolyzer could be set to a constant value, up to a maximum level, owing to the hydrogen storage pressure or the minimum applied load.
The linear fit of the average electrolyzer cell-voltage shows an increase of 0.5 µV/h during the testing period. That is better than the reported literature values shown on ***Figure 5***. The other reported degradation rates span a wide range between 1.5 µV/h and 230 µV/h.[5]
It should be mentioned that the applied current density of our electrolyzer during the testing intervals is in the range of 0.25 – 1 A/cm^2. The results from ***Figure 5*** are obtained at a current density of 0.5 A/ cm^2.
Assuming a similar linear trend in the cell voltage decay and similar operating conditions and patters, the electrolyzer would reach a 70 % average cell efficiency (at a 100 % load) after 1.2 million operating hours. A significantly longer testing period would be needed to validate this assumption.

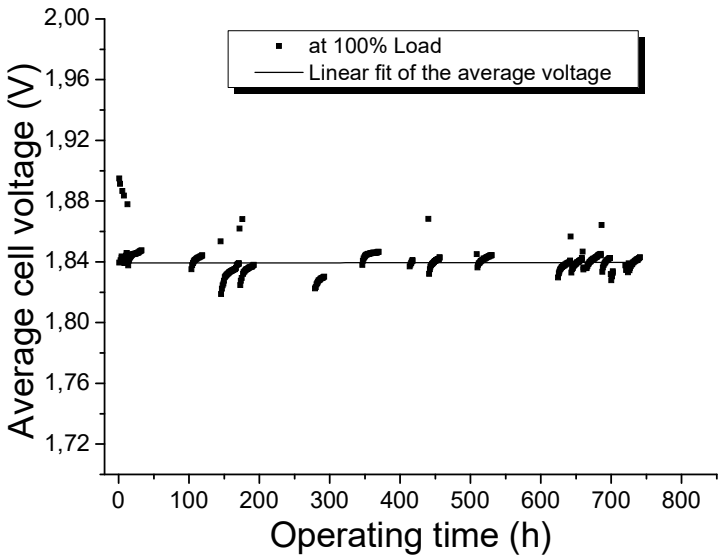

Figure 4. The recorded variation of the hourly average electrolyzer cell voltage as a function of the operating time.

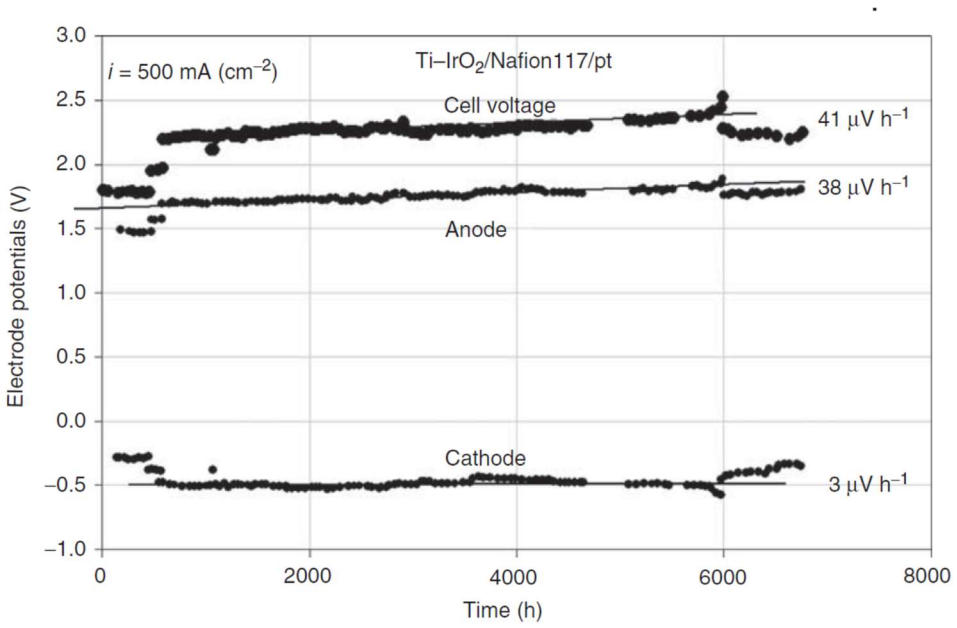

Figure 5. Example of a reported literature electrolyzer cell voltage decay.[6]

The change of the measured system efficiency of the electrolyzer is in direct contradiction with the cell voltage results (**Figure *6***). The system efficiency was seen to drop significantly during the testing period. The original data shows that this efficiency drop is due to the decreased hydrogen production. The regularly executed regeneration of the drying columns is also causing some hours (the lagging data points) with a decreased system efficiency.

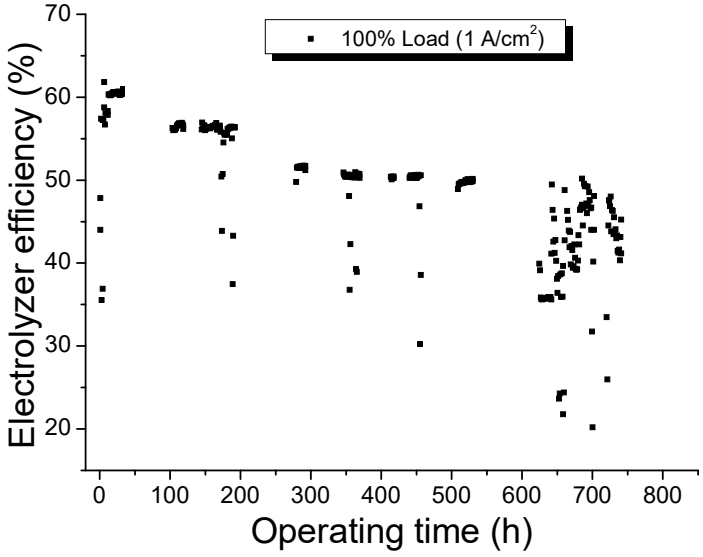

Figure 6. The variation of the electrolyzer system efficiency as a function of the operating time.

The stable cell efficiencies suggest that the amount of produced hydrogen is not changing in reality, but some hydrogen leaks or a more likely, a flowmeter error could cause these

(mis)readings. At the beginning of the lifetime of the experimental setup, a malfunctioning check valve allowed the direct flow of wet hydrogen into the hydrogen storage tank. This high amount of moisture had a detrimental effect on the Bronkhorst flowmeter. By drying the flowmeter the issue was seemingly solved, but it is possible that this incident caused a gradual device deterioration.

PEM fuel cell

The average cell voltage of the fuel cell dropped with a rate of 2 µV/h during the studied time period. A decreasing cell voltage is a sign of a decreased cell efficiency of a fuel cell. It should be noted in **Figure** *7* that similarly to the electrolyzer cell, when the fuel cell is continuously operated for several hours, the cell voltage drops significantly. After a break (in the weekend for example) the cell voltage is shifted back almost to the original level. Membrane humidity level or proton depletion of the membrane could explain this behaviour. However, on a longer timescale, the average cell voltage is decreasing as is shown by the red fitted line.
The cell efficiency is decreasing with a similar rate (**Figure *8***), which is a direct consequence of the calculation method of the cell efficiency.

The 2 µV/h deterioration rate of the fuel cell under similar operating conditions would allow 140 000 hours total operating time before the cell efficiency would drop below 30 %.

The fuel cell system efficiency is dropping with a rate similar to the cell efficiencies, as is shown in **Figure *9***.

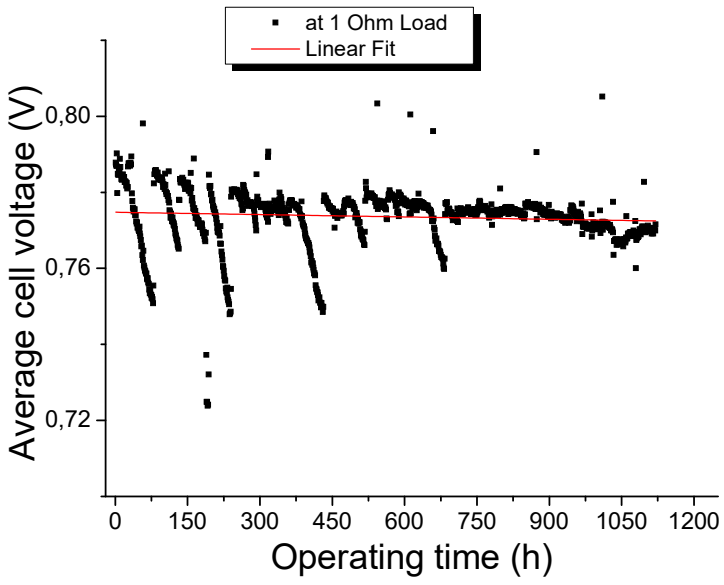

Figure 7. The recorded variation of the hourly average cell voltage of the studied PEM fuel cell as a function of the operating time.

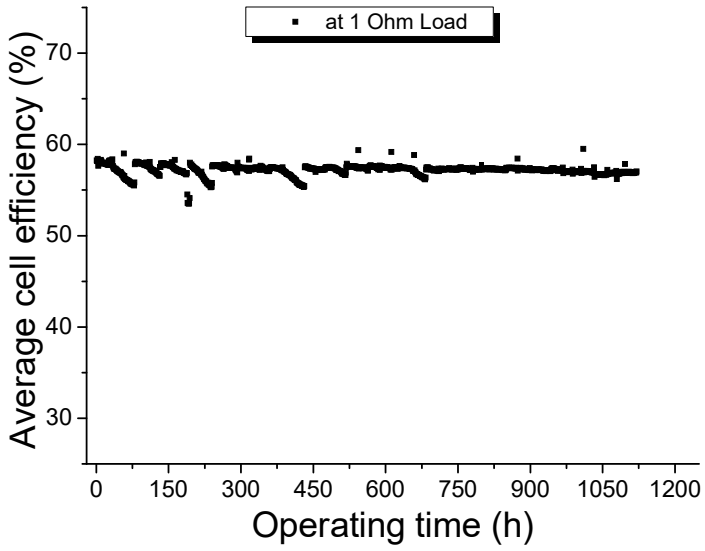

Figure 8. The calculated average hourly cell efficiencies of the PEM fuel cell plotted as a function of the operating time.

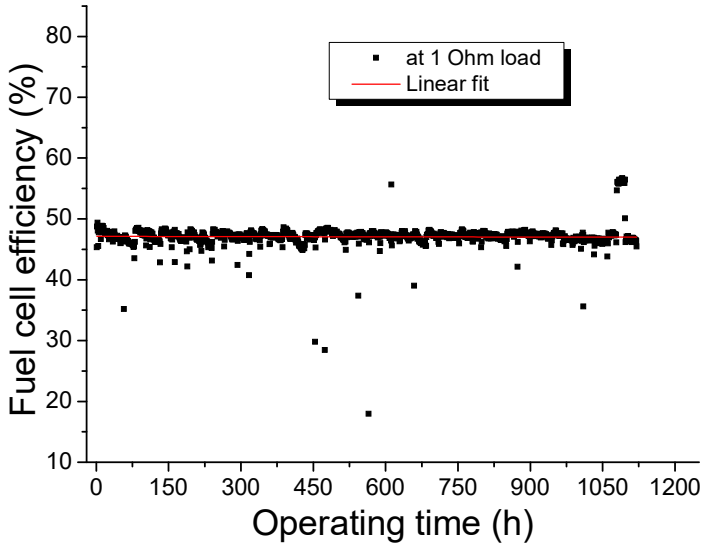

Figure 9. The calculated average hourly fuel cell efficiencies shown as a function of the operating time.

7. Conclusions

The electrolyzer unit was operated at various loads and cell pressures for a total of about 750 hours to investigate long-term stability. The electrolyzer cell showed a stable cell voltage and cell efficiency in the studied time period, with an average cell voltage decay rate of 0.5 µV/h. The apparent system efficiency of the electrolyzer showed however a significant drop, which contradicts the cell efficiency results. The reason for this efficiency drop is still unknown, but a malfunctioning flow meter is suspected to cause these results. Assuming a similar linear trend in the cell voltage decay and similar operating conditions and patters in the future, the obtained voltage decay rate suggests a several hundred thousand of hours or even longer expected electrolyzer stack lifetime. A significantly longer testing period would be needed to validate this results-based assumption.

The fuel cell was operated at a constant load of 1 Ω resistance for a total of about 1120 hours to investigate long-term stability. The average cell voltage of the fuel cell dropped with a rate of 2 µV/h during the studied time period. Both the cell efficiency and system efficiency of the fuel cell showed a comparable decreasing trend. Under similar operating conditions the measured cell voltage decay rate would allow 140 000 hours of further operating time before the cell efficiency would drop below 30 %.

8. Acknowledgement

Financial support from GasTerra and GasUnie is acknowledged.

9. References

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